Final Technical Report for the Project: *"Synthesis and Magnetic, Thermal, and Electrical Measurements on Complex non-Cuprate Superconductors"*.
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Laboratory Partnership Abstract

The project investigated superconductivity in non-cuprate materials with critical temperatures, T_c, in excess of 20 K in order to understand the thermodynamics of several of these materials. The project is a cooperative effort between investigators at Southern University (SU), Louisiana State University (LSU), and Los Alamos National Laboratory (LANL). It involved synthesis of high quality samples, and subsequent detailed magnetic, thermal and electrical measurements on them. The project provided a PhD Thesis research experience and training for a graduate student, Ms. Robin Macaluso. High quality, single crystal samples were synthesized by Ms. Macaluso under the direction of one of the CO-PIs, John Sarao, during the summer while she was a visitor at LANL being supported by this grant. On these samples magnetic measurements were performed at SU, thermal and electrical measurements were made in the LSU Physics and Astronomy Department. The crystallographic properties were determined in the LSU Chemistry Department by Ms. Macaluso under the direction of her dissertation advisor, Dr. Julia Chan. Additional high field magnetic measurements on other samples were performed at the National High Magnetic Field Laboratory (NHMFL) both in Tallahassee and at LANL. These measurements involved another graduate student, Umit Alver, who used some of the measurements as part of his PhD dissertation in Physics at LSU.

DISCLAIMER

Summary of Accomplishments

The following is a partial summary of the major accomplishments from the project.

(1). Crystal growth.

High quality single crystals of $Ba_{1-x}K_xBiO_3$ and $Ba_{1-x}Rb_xBiO_3$ (x = 0.4), and $Ba_{0.25}Rb_{0.25}BiO_3$ were grown electrochemically using a three-electrode crystallization method. Fig. 1 shows a schematic of the experimental setup developed during the grant period.

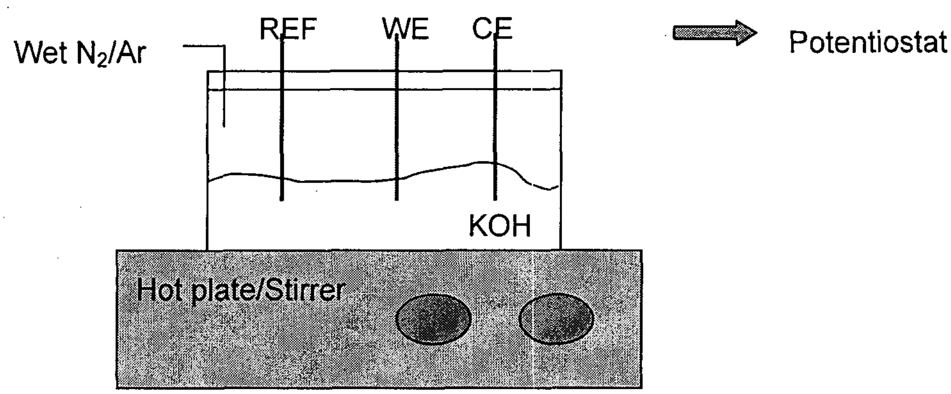


Fig. 1

Typical crystals produced possess cubic morphology, reflecting the cubic crystal structure (Pm3m space group). The crystals are large and individual cubes are visible with the naked eye.

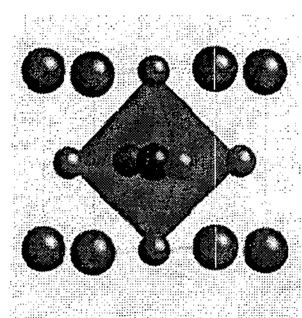


Fig. 2 The Pm3m space group

SQUID magnetization measurements of the $Ba_{0.25}Rb_{0.25}BiO_3$ showed a superconducting transition temperature close to 26 K.

(2) X-ray structure determinations.

Structure of $Ba_{0.6}K_{0.4}BiO_3$ determined from x-rays at LSU.

A sample of $Ba_{0.6}K_{0.4}BiO_3$ underwent resonant ultrasound measurements as a function of temperature at Los Alamos National Laboratory. The initial cooling measurements showed a surprising result that there is a first order hysteretic phase transition in this cubic perovskite high T_c material near 200 K that gives rise to a large change in both the sound attenuation and frequency dispersion. Detailed x-ray structure determinations as a function of temperature were carried out through this new transition.

(3) Magnetic and thermal measurements.

There is an ongoing question of the thermodynamic order of the superconducting transition in $Ba_{0.6}K_{0.4}BiO_3$ (BKBO). We investigated alloys of BKBO and $Ba_{0.75}Rb_{0.25}BiO_3$ in an attempt to lower T_c to a point where the phonon contribution to the specific heat is much smaller without changing the crystallographic structure of the material. Results from one of the samples of $Ba_{0.75}Rb_{0.25}BiO_3$ show the superconducting transition in the magnetization, but not in the specific heat. (Figures 3 and 4) BKBO show similar results.

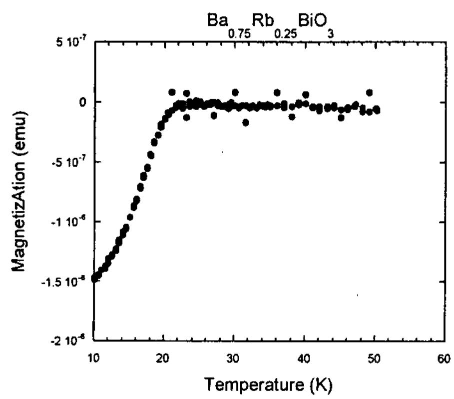


Figure 3. Magnetization vs. Temperature of single crystal$Ba_{0.75}Rb_{0.25}BiO_3$ showing the superconducting transition.

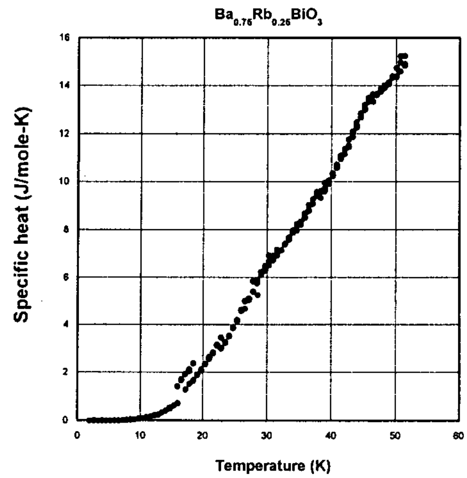
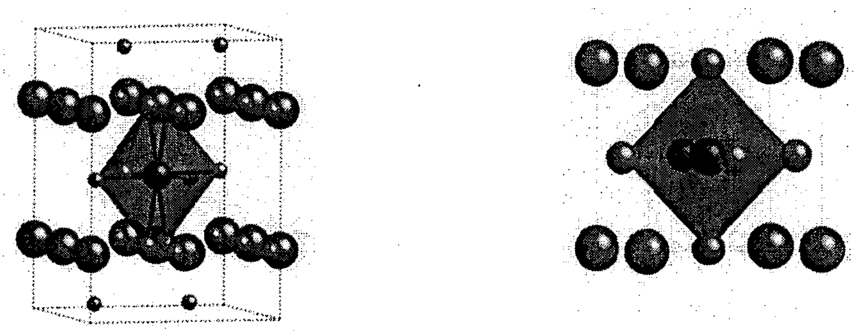


Figure 4. Specific heat vs. Temperature of single crystal $Ba_{0.75}Rb_{0.25}BiO_3$ showing practically no structure near the superconducting transition at 20 K.

(4) Temperature dependence of the crystallographic parameters.

Structurally $Ba_{0.6}K_{0.4}BiO_3$ (BKBO) is interesting because no CuO_2 planes exist to mediate superconductivity. BKBO has the highest T_c (31 K at x = 0.4) in this family of compounds. A related superconducting compound (T_c = 13 K), $Ba_{1-x}Pb_xBiO_3$, also has a cubic perovskite structure. The crystal structure of $BaPbBiO_3$ is known to undergo transformations from orthorhombic → tetragonal → orthorhombic → monoclinic as a function of x and superconductivity is confined to the tetragonal phase.

Published results of neutron diffraction studies on single-crystal BKBO indicate an *I4/mcm* space group at 50 K. Examination by powder diffraction technique on crushed single crystals using synchrotron radiation show the same space group. At T = 300 K powder neutron diffraction data shows the compound is in *Ibmm* space group. The crystal structures of *Ibmm* and *Pm3m* space groups are shown in Figures 5a and 5b.



(a) (b)

Figure 5. BKBO in the *bmm* space group (a) and the *m3m* space group (b).

We carried out single-crystal X-ray diffraction on a BKBO crystal, and our thermal parameters are consistent with published results. However, our refined model fits best with the Pm3m space group at temperatures between 110 K and 300 K. Lattice parameters and R_w values are given in Table1.

Table 1.

Temperature (K)	Lattice Parameter (Å)	R (%)
110	4.2729	5.7
150	4.2800	
175	4.2775	
200	4.2800	
225	4.2797	
250	4.2802	
299	4.2826	4.6

The oxygen thermal parameters (U_{33}) have been measured at various temperatures in an attempt to identify any octahedral tilts and/or distortions. The thermal parameters measured are compared to those measured by Braden et al. in Figure 6.

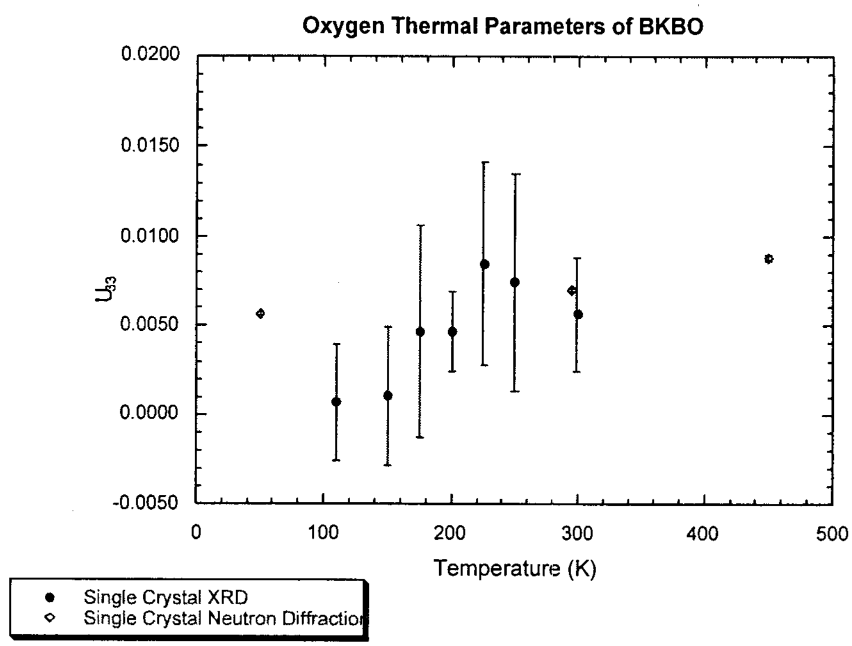


Figure 6. Braden's data in green circles. In that neutron analysis the thermal parameters of oxygen were constrained to be identical.

(5) Other materials.

We synthesized $BeB_{2.75}$ and measured the electron transport properties. Our sample is superconducting below $T_c = 0.72$ K with a critical field of $H_{c2} = 0.175$ T. See Fig. 7.

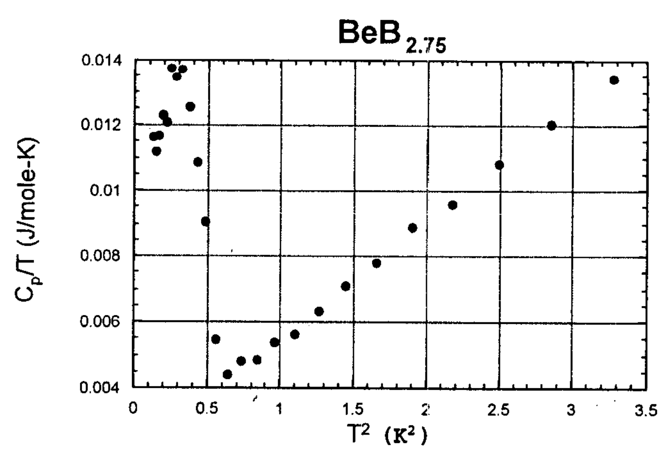


Figure 7. Specific heat divided by Temperature vs. Temperature squared showing that the superconducting transition near 0.7 K is clearly observed.

(6) In addition to the work on which we have reported at the end of each reporting period, and described above, considerable effort was made in crystal growth (by Ms. Macaluso at LANL) and characterization of a new set of highly correlated superconducting and magnet materials: (Ce,La)MIn_5 where M = Co, Ir, or Rh, known as the 115s. The funds provided by the grant have led to many successful cooperative projects on the 115s and other superconductors between one of the principal investigators, Roy G. Goodrich and the group led by another of the principal investigator at LANL, John L. Sarrao. Other collaborations between Goodrich and groups at LSU also were productive. The papers resulting from work during the award period are listed below in reverse chronological order.

Publications and collaborations.

1. Harrison, N., Alver, U., Goodrich, R. G. et al., "4f-electron localization in $Ce_xLa_{1-x}MIn_5$ with M = Co, Rh, or Ir", Phys. Rev. Lett., **93**, 186405 (2004).

2. Goodrich, R. G., Browne, D, Kurtz, R., et al, "de Haas-van Alphen measurements of the electronic structure of $LaSb_2$", Phys. Rev. B, **69** (12): Art. No. 125114 MAR 2004.

3. Goodrich, R. G., Young, D. P., Hall, D., et al., Extension of the temperature-magnetic field phase diagram of CeB_6. Phys. Rev. B, **69** (5): Art. No. 054415 FEB 2004.

4. Movshovich, R., Bianchi, A., Capan, C., et al. "Electron-spin domains - magnetic enhancement of superconductivity". Nature, **427** (6977): 802-802 FEB 26 2004.

5. Young, D. P., Goodrich, R. G., Ditusa, J. F., et al. High magnetic field sensor using $LaSb_2$. Appl. Phys. Lett., **82** (21): 3713-3715 MAY 26 2003.

6. Young, D. P., Fisk, Z., Thompson, J. D., et al. magnetic properties - parasitic ferromagnetism in a hexaboride? Reply. NATURE **420** (6912): 144-144 NOV 2002
7. Macalouso, R. T., Sarrao, J. L. Pagliuso, P. G., et al. Crystal growth and structure determination of $LaMIn_5$ (M = Co, Rh, Ir). J. Solid State. Chemistry, **166** (1): 245-250 JUN 2002.

8. Young, D. P., Goodrich R. G., Adams, P. W., et al. Superconducting properties of $BeB_{2.75}$ Phys. Rev. B **65** (18): Art No. 180518 MAY 1 2002.

9. Murphy, T. P., Hall, D., Palm, E. C., et al. Anomalous superconductivity and field induced magnetism in $CeCoIn_5$. Phys. Rev. B, **65** (10): Art. No. 100514 MAR 1 2002.

Number of Undergraduates Supported by DOE
None (0)
Number of Graduates Students Supported by DOE
One (1)
Number of Postdoctoral Fellows Supported by DOE
None (0)